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BRINGING TOGETHER COMPUTATIONAL AND EXPERIMENTAL CAPABILITIES AT THE CRYSTAL SCALE

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Abstract. Many phenomena of interest occur at the scale of crystals or are controlled by events happening at the crystalline scale. Examples include allotropic phase transformations in metals and pore collapse in energetic crystals. The research community is increasingly able to make detailed experimental observations at the crystalline scale and to inform crystal scale models using lower length scale computational tools. *In situ* diffraction techniques are pushing toward finer spatial and temporal resolution. Molecular and dislocation dynamics calculations are now able to directly inform mechanisms at the crystalline scale. Taken together, these factors give crystal based continuum models the ability to rationalize experimental observations, investigate competition among physical processes, and, when appropriately formulated and calibrated, predict behaviors. We will present an overview of current efforts, with emphasis on recent work investigating phase transformations and twinning in metals. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344 (LLNL-CONF-415155).

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INTRODUCTION

Phase transformation and twinning are of active interest across a variety of material systems, for example [1, 2, 3, 4, 5]. Understanding of these deformation mechanisms is often sought at the crystal scale and involves interaction with other mechanisms such as crystallographic slip by dislocation glide. Phase transformation and twinning pose several modeling challenges including the fact that they involve large strains producing material with new crystal lattice orientation. Here we will discuss one model for simulating phase transformation and twinning at the crystal scale and for making connections to experiments focused at the same scale.

Many other perspectives and modeling approaches have been proposed, with a small sampling of these being [6, 7, 8, 9, 10, 11, 12, 13, 14, 15]. The approach to phase transformation and twinning used here is somewhat unique in that it is a crystal mechanics

based approach which includes elastic and plastic accommodation, is applicable to large strains including large volume changes and to complete transformation, and is formulated to work for either quasi-static loading or dynamic loading scenarios. As in some other recent approaches [16, 17], we include the effects of anisotropic elasticity on the driving forces for phase transformation and twinning.

Details of the formulation are contained in [18, 19] and [20] discusses slip and dislocation multiplication kinetics effects for a related model. Mass fractions track the amounts of parent and twinned/transformed domains in a material element. Material within each domain is allowed to deform according to anisotropic crystallographic slip systems and anisotropic thermo-elastic behavior associated with its specific lattice orientation. Twinning/transformation produces mass fraction rates among the microstructural constituents. The response of each material point is then the collective

response of all of the constituent responses combined with the available twinning/transformation modes. Solution of the constitutive update problem proceeds by forming a residual on satisfaction of the overall kinematics and the kinetics for mass fraction transfer rates. Constraints of various sorts enforce temperature and stress equilibrium, mass conservation, and admissible mass fractions.

The versatility of the model used here makes it useful for connecting to a variety of experiments at the crystal scale and for exploring the implications of various modeling assumptions. For example, researchers are actively exploring modeling assumptions for dislocation kinetics [20, 21, 22]. One avenue of exploration in the twinning/transformation context is a phenomenological interaction energy. This energy, typically a quadratic form in the mass fractions, may be used to penalize the coexisting of certain types of microstructural constituents. These forms may be motivated by surface energy or storage of elastic energy from incompatibility effects at the subscale. As shown below, this interaction energy can be used to promote the formation of relatively sharp twin domains, but more physically motivated forms, perhaps with anisotropic surface energy effects, may ultimately be required for proper resolution of domain structure.

COMPLEXITIES ASSOCIATED WITH THE $\alpha \leftrightarrow \epsilon$ TRANSFORMATION IN IRON

Results in this section use essentially the same formulation and parameters as shown in [18] for the $\alpha \leftrightarrow \epsilon$ transformation in iron and here we explore complexities associated with the multiplicity of deformation mechanisms. As one example, we can simulate the quasi-static deformation of an iron polycrystal through the phase transformation, as shown in Figure 1. We see several interesting features: initiation of transformation at stress concentrations near grain boundaries; variant selection based on local deviatoric stress state; and cooperative transformation across many grains in bands roughly perpendicular to the loading direction.

Features of this prediction depend on model parameters, and these parameters are often difficult to calibrate using macroscopic observables. For example, observed pressure rise during transforma-

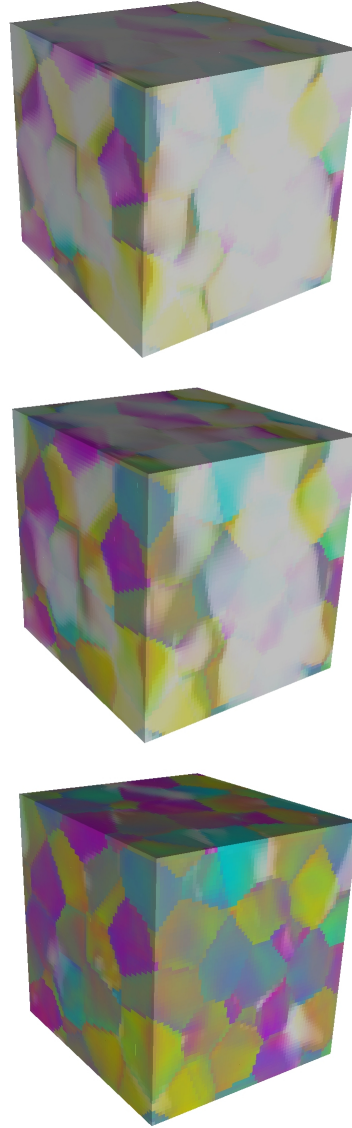


FIGURE 1. Iron polycrystal quasi-statically compressed through the α to ϵ transformation following the Burgers mechanism. Colors indicate ϵ variant selection, with white/grey indicating α phase.

tion [23] may be predicted by either increasing interaction energy constants or increasing the resistance to transformation produced by dislocations in the material which accumulate as the transformation progresses. Calibration choices affect predicted microstructures and change the results predicted under

other loading scenarios. Overall, work on iron and quartz [18, 19, 5] shows that the model can reproduce several experimentally observed quantities, and indicates that connecting to grain-resolved experiments would be better for validating and calibrating the model in a way which improves subsequent predictive capability.

Laser driven experiments on (100) oriented iron single crystals provide one source of validation data [24]. These experiments have the interesting property that they are on a time scale short enough to suppress plasticity by dislocation glide, as confirmed by both *in situ* observations and detailed examination of recovered samples. This allows deviatoric stresses to be higher, which influences variant selection. The Bain path is used for the transformation mechanism and, upon reverse transformation, the model allows the material in each ϵ variant to return to the parent α variant or to two other α variants allowed under the mechanism and symmetry of the ϵ phase. The non-parent α variant predictions are in very good agreement with data from recovered samples only if the relaxation of deviatoric stresses by dislocation glide is effectively eliminated over the time scale of the simulation. This provides a nice validation of the kinematic pathways, but does not on its own particularly constrain any parameters in the kinetics of slip or transformation. In part this is because the level of detail currently available from dynamic experiments remains limited. Furthermore, extensive shot campaigns for experiments of this sort are not undertaken lightly.

Quasi-static studies focused on single crystals are possible, but single crystals are hard to obtain for many materials of interest. Even when single crystals can be obtained, it is challenging to deform them in the manner desired [25], and their behavior may be different from that exhibited in bulk polycrystalline samples.

GRAIN RESOLVED SIMULATION AND EXPERIMENTAL OBSERVATIONS OF TWINNING

One path forward is to use new X-ray diffraction based techniques for obtaining detailed data at the crystalline grain scale from bulk polycrystals. Preliminary results along these lines have been pre-

sented for twinning in a magnesium alloy [26], and newer techniques speed up collection times making the method attractive for *in situ* quasi-static studies. For each grain, the method provides average tensorial stretch of the lattice, relative spatial position, and lattice orientation. Relative intensities provide information about volume fractions of twinned/transformed domains.

While the preliminary data in [26] are not rich enough for a direct calibration, the results raise some interesting issues for use of the data in model calibration. It is shown that in the average over the diffraction volume, the twins and the parent grain do not exhibit traction balance across the habit plane. This points to substantial heterogeneities over the grain. We therefore wish to determine whether such heterogeneities can be captured in a model which treats other anisotropic elastic and plastic accommodation effects in three dimensions.

A rough model calibration uses stress/strain response and bulk diffraction data for evolving lattice orientation distributions due to twinning [27], and parameters from previous crystal level modeling efforts [28].

As the interaction energy in the model is increased, finite element simulations discretizing grains of a polycrystal simulate sharp twin formation and pop-in events wherein twins appear abruptly accompanied by stress drops. These simulations use an implicit-dynamic formulation to stabilize the solution. Figure 2 shows the change in predicted twin morphology with increasing interaction energy for an idealized $5 \times 5 \times 5$ grain polycrystal at relatively coarse mesh refinement. Interaction energy promotes localized twinning and long range interactions through the stress field promote planar twin domains. Twin events in one grain can trigger twinning in adjacent grains due to stress concentrations. Figure 3 shows two views of a grain at higher mesh resolution from a polycrystal simulation. A single twin variant dominates the grain and it tends to form planar domains, but two habit planes are formed instead of the one which is expected from typical experimental observations of twinning in metals. It is worth noting that the position of the twins is determined by the simulation and is not specified in advance – changing the neighborhood of the grain changes the twin locations and can even alter variant selection.

Following the method used by Ericksen [29] we

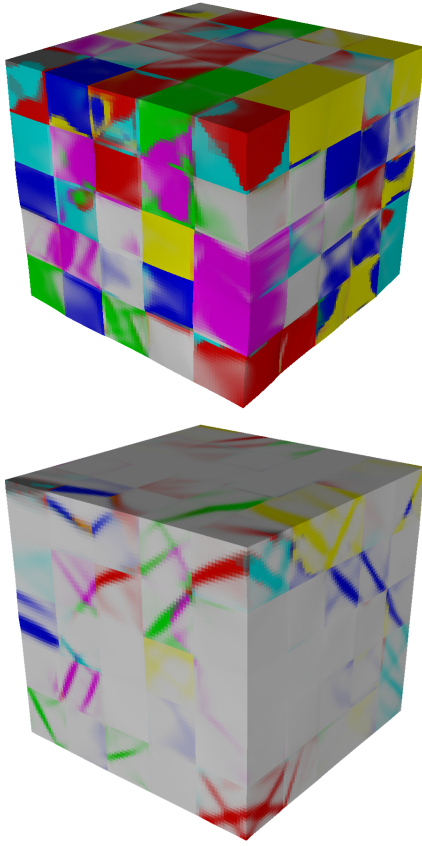


FIGURE 2. Twin morphology for a polycrystal, at relatively low (top) and high (bottom) values of interaction energy. Mass fractions for the six twin variants are mapped to red, green, blue, yellow, cyan, and magenta.

see that indeed the analytic solution of the habit plane orientation has two solutions. This analysis proceeds by considering the Hadamard jump condition which requires the difference in deformation gradients on either side of the interface to be a rank-one tensor, or $[\mathbf{F}] = \bar{\mathbf{a}} \otimes \bar{\mathbf{n}}$. Neglecting all deformation other than the given twinning mode, we have $\mathbf{F}_- = \mathbf{I}$ and $\mathbf{F}_+ = \mathbf{I} + \gamma \mathbf{s} \otimes \mathbf{m}$. For twinning in the magnesium alloy of interest, $\gamma \approx 0.129$ and the two solutions for $\bar{\mathbf{n}}$ fall at 0° and 86.3° from \mathbf{m} in the plane spanned by \mathbf{s} and \mathbf{m} . The graphical solution is shown in Figure 4 with the untwinned shape in blue and the twinned shape overlaid. These two solutions are used to draw the slicing planes shown in Figure 3. Real materials exhibit only the 0° solution because that is the solution for a coherent twin which requires relatively

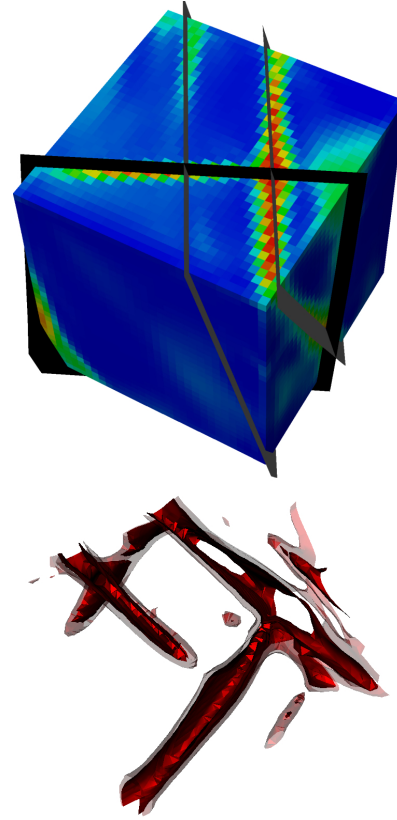


FIGURE 3. Twin morphology for a grain within a deformed polycrystal. Calculated habit planes are shown (top) and isosurfaces (bottom, from a different view) show that structure is planar through the whole of the grain.

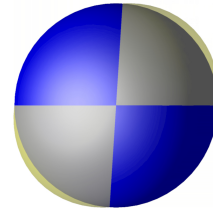


FIGURE 4. Graphical depiction of habit plane solutions for $\gamma = 0.129$.

few dislocations to accommodate the boundary. The other solution would have substantially higher surface energy, and may also be less favored due to growth kinetics.

In future work, it may be possible to include the

effects of anisotropic surface energy through the gradients in variant mass fractions. Interactions between twins and dislocation populations are actively being investigated [30] and energy storage associated with dislocation structures depends in some detail on the character of the structures formed [31].

Note that while the grain reported in the experimental work [26] exhibits two twin variants, simulations to date of a grain at that same orientation produce just a single variant. In the model, the driving force for a second variant is a substantial fraction of the force for the twin which is formed, but the second twin does not form due to highly nonlinear kinetics. Differences could be due to improper model calibration, neighborhood effects, or improper twin morphology predictions. Further investigation using a more detailed experimental data set might allow a more definitive understanding.

DIRECTLY MEASURING STRENGTH OF DEFORMATION MECHANISMS

Preliminary data from an α titanium alloy, shown in Figure 5, provides another example of how crystal level models may connect directly to diffraction experiments. In this data, the tensorial lattice stretches are measured in a handful of grains as the load is increased. These stretches are converted to stresses which may then be projected onto slip systems. The values for basal and prismatic slip in this HCP material plateau with increasing macroscopic load while the values for pyramidal slip continue to rise, indicating that pyramidal slip is more difficult to activate. With faster collected times and the ability to collect data from more grains, these types of measurements may directly provide strength determinations for specific deformation mechanisms in materials which are difficult to access, such as the HCP ϵ phase in iron.

CONCLUSIONS

Prospects are good for validation and calibration of models phrased at the crystal scale. In some cases, it is possible for experiments to isolate particular mechanisms. When this is not possible, novel X-ray diffraction experiments which provide unprecedented levels of detail at the crystal scale (relative

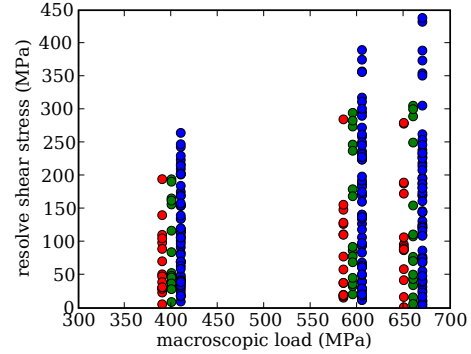


FIGURE 5. Preliminary data showing projection of grain-specific stresses onto basal (red), prismatic (green), and pyramidal-1 ($c + a$) (blue) slip systems for grains in an α -titanium (HCP) alloy.

position and volume fractions as well as full tensorial lattice strain) offer a path forward. With further model development, one may be able to capture twin morphology in a meaningful manner in real material systems. This could allow for more direct connection to experimental results. Various techniques are pushing to finer three dimensional diffraction based measurement [32], though so far with measurement times which preclude many *in situ* loading scenarios.

Heterogeneities associated with twinning and phase transformations will continue to pose modeling and experimental challenges for some time to come. Substantial advances have been made both in directly resolving heterogeneities and in homogenization schemes which account for some of the effects of heterogeneity [33, 34, 35]. For use in component scale simulations, schemes which are based on non-local evolution of probability densities over orientation space [36, 37] hold some promise for efficient multi-scale embedding such as the sort proposed in [38, 39]. Experimental and modeling work focused at the crystal scale can help to inform such homogenized methods.

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REFERENCES

1. Merkel, S., Wenk, H.-R., Gillet, P., Kwang Mao, H., and Hemley, R. J., *Physics of the Earth and Planetary Interiors*, **154**, 239–251 (2004).
2. Blumenthal, W. R., Brown, D. W., and Tomé, C. N., “Evolution of Crystallographic Texture and Strength in Beryllium,” in *Shock Compression of Condensed Matter - 2003: Proceedings of the Conference of the American Physical Society Topical Group on Shock Compression of Condensed Matter*, AIP, 2004, vol. 706, pp. 525–528.
3. Field, R., McCabe, R., Alexander, D., and Teter, D., *Journal of Nuclear Materials*, **392**, 105 – 113 (2009).
4. McNaney, J. M., Barton, L. M. H. N. R., and Kumar, M., “Shock driven twinning in tantalum single crystals,” in *16th International Conference on Shock Compression of Condensed Matter*, 2009.
5. Wenk, H.-R., Barton, N., Bortolotti, M., Vogel, S., Voltolini, M., Lloyd, G., and Gonzalez, G., *Physics and Chemistry of Minerals* (2009), in press.
6. Duvall, G. E., and Graham, R. A., *Reviews of Modern Physics*, **49**, 523–579 (1977).
7. Abeyaratne, R., and Knowles, J. K., *J. Mech. Phys. Solids*, **38**, 345–360 (1990).
8. Abeyaratne, R., Chu, C., and James, R. D., *Phil. Mag. A*, **73**, 457–497 (1996).
9. Boettger, J. C., and Wallace, D. C., *Phys. Rev. B*, **55**, 2840–2849 (1997).
10. Diani, J. M., and Parks, D. M., *J. Mech. Phys. Solids*, **46**, 1613–1635 (1998).
11. Rengarajan, G., and Reddy, J. N., *J. Mech. Phys. Solids*, **49**, 1665–1700 (2001).
12. Artemev, A., Jin, Y., and Khachaturyan, A. G., *Acta Mater.*, **49**, 1165–1177 (2001).
13. Levitas, V. I., Idesman, A. V., Olson, G. B., and Stein, E. N., *Phil. Mag. A*, **82**, 429–462 (2002).
14. Yano, K., and Horie, Y., *Int. J. Plasticity*, **18**, 1427–1446 (2002).
15. Anand, L., and Gurtin, M. E., *J. Mech. Phys. Solids*, **51**, 1015–1058 (2003).
16. Zhang, W., Jin, Y. M., and Khachaturyan, A. G., *Phil. Mag.*, **87**, 1545–1563 (2007).
17. Zhang, R. Y., Daymond, M. R., and Holt, R. A., *Mater. Sci. Eng., A*, **473**, 139 – 146 (2008).
18. Barton, N. R., Benson, D. J., and Becker, R., *Modelling Simul. Mater. Sci. Eng.*, **13**, 707–731 (2005).
19. Barton, N. R., and Wenk, H.-R., *Modelling Simul. Mater. Sci. Eng.*, **15**, 369–394 (2007).
20. Barton, N. R., Winter, N. W., and Reaugh, J. E., *Modelling Simul. Mater. Sci. Eng.*, **17**, 035003 (2009).
21. Armstrong, R. W., and and. F. J. Zerilli, W. A., *J. Appl. Phys.* (2009).
22. Devincere, B., Hoc, T., and Kubin, L., *Science*, **320**, 1745–1748 (2008).
23. von Bargen, N., and Boehler, R., *High Pressure Res.*, **6**, 133–140 (1990).
24. Hawreliak, J., Colvin, J. D., Eggert, J. H., Kalantar, D. H., Lorenzana, H. E., Stolken, J. S., Davies, H. M., Germann, T. C., Holian, B. L., Kadau, K., Lomdahl, P. S., Higginbotham, A., Rosolankova, K., Sheppard, J., and Wark, J. S., *Phys. Rev. B*, **74**, 184107 (2006).
25. Rae, P. J., Hooks, D. E., and Liu, C., “The stress versus strain response of single β -hmx crystals in quasi-static compression,” in *Proceedings of the Thirteenth International Detonation Symposium*, Office of Naval Research, 2006.
26. Aydiner, C. C., Bernier, J. V., Clausen, B., Lienert, U., Tomé, C. N., and Brown, D. W., *Phys. Rev. B* (2009), in press.
27. Clausen, B., Tomé, C., Brown, D., and Agnew, S., *Acta Mater.*, **56**, 2456 – 2468 (2008).
28. Staroselsky, A., and Anand, L., *Int. J. Plasticity*, **19**, 1843–1864 (2003).
29. Ericksen, J. L., *Arch. Rational Mech. Anal.*, **88**, 337–345 (1985).
30. Kochmann, D. M., and Le, K. C., *J. Mech. Phys. Solids*, **57**, 987–1002 (2009).
31. Benzerga, A., Bréchet, Y., Needleman, A., and Van der Giessen, E., *Acta Mater.*, **53**, 4765 – 4779 (2005).
32. Yang, W., Larson, B. C., Tischler, J. Z., Ice, G. E., Budai, J. D., and Liu, W., *Micron*, **35**, 431–439 (2004).
33. Proust, G., Tomé, C. N., and Kaschner, G. C., *Acta Mater.* (2007).
34. Proust, G., Tomé, C. N., Jain, A., and Agnew, S. R., *Int. J. Plasticity*, **25**, 861–880 (2009).
35. Lebensohn, R., *Modelling Simul. Mater. Sci. Eng.*, **7**, 739–746 (1999).
36. Lebensohn, R. A., and Tomé, C. N., *Mater. Sci. Eng.*, **A175**, 71–82 (1994).
37. Myagchilov, S., and Dawson, P. R., *Modelling Simul. Mater. Sci. Eng.*, **7**, 975–1004 (1999).
38. Knap, J., Barton, N. R., Hornung, R. D., Arsenlis, A., Becker, R., and Jefferson, D. R., *Int. J. Num. Meth. Eng.*, **76**, 572–600 (2008).
39. Barton, N. R., Knap, J., Arsenlis, A., Becker, R., Hornung, R. D., and Jefferson, D. R., *Int. J. Plasticity*, **24**, 242–266 (2008).